PATENT SPECIFICATION

DRAWINGS ATTACHED

893,041

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COMPLETE SPECIFICATION

Process for the Manufacture of Shaped Bodies of Silicon Carbide

We, WACKER-CHEMIE G.M.B.H., a Company recognised by German law, of 22, Prinzregentenstrasse, Munich 22, Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The commercial methods for the production of silicon carbide lead to a product which adequately fulfils the ordinary uses of silicon carbide. However, the product is not entirely suitable for use as a semi-conductor, for which purpose certain electrical and optical or crystallographic properties are required, and especially when the silicon carbide must have a certain well defined shape. It is difficult to produce the usual silicon carbide in the form of pieces of well defined shape.

The present invention is based on the observation that highly pure silicon carbide can be produced in the form of bodies of any desired shape by heating a shaped body of graphite which may contain silicon carbide in a gaseous atmosphere comprising a silicon subhalide. There may be used as starting material commercial silicon carbide from which silicon is at least partially removed or purified and shaped graphite. When commercial silicon carbide is used as starting material the pieces, which are generally of ill defined shape, are shaped by breaking or grinding to form small plates, rods or discs. As graphite can easily be worked up into hollow bodies, the process of the invention enables complex bodies such as tubes or vessels of very pure silicon carbide to be made in a simple manner.

The bodies made of commercial silicon carbide are wholly or partially freed from silicon by treatment with chlorine or a silicon tetrahalide at a temperature above 1000° C. In this treatment all the harmful impurities are simultaneously removed from the crystal structure. When the whole of the silicon is removed, there remains behind very pure graphite having the habit of the former silicon carbide body. The highly pure body which is wholly or partially free from silicon is then siliconised in a gaseous atmosphere until the formation of silicon carbide is complete. This may be carried out in the following way:—

A silicon subhalide, for example silicon dichloride, is produced from silicon and silicon tetrachloride, and the resulting dihalide, if desired in the presence of hydrogen, is disproportionated into silicon and silicon tetrahalide on the graphite body, whereupon the silicon reacts with the graphite body to form silicon carbide. The process is advantageously carried out at a temperature in the range of 1300°—1400° C.

The method is illustrated with reference to the accompanying drawings.

The treatment is carried out as shown in Fig. 1 in attached drawings in a non-isothermic reaction vessel 2, in which is housed within the heating element 1. At one end of the vessel is located the purified graphite body 4 at a lower temperature T_1 than the highly purified silicon 3 having a temperature T_2 and located at a distance from the body 4. Silicon tetrahalide is advantageously used as the medium for transferring the silicon 3 to the graphite body 4. At the hotter part of the vessel gaseous silicon dichloride is formed from silicon, and, for example, silicon tetrachloride vapour, and the gaseous silicon dichloride passes by convection and diffusion to the colder graphite body, where it decomposes into silicon and silicon tetrachloride. The silicon tetrahalide recovered then forms fresh silicon dichloride by reaction with the hot silicon 3.

The temperature diagram in Fig. 2 shows

that the body to be siliconised 4 is maintained at a lower temperature than the body 3. The temperature T_1 is higher than T_2.

The precipitated silicon reacts immediately with the graphite body to form silicon carbide. When the process is carried out in a quartz tube the siliconisation can be closely observed and discontinued at the instance when the formation of silicon carbide is complete. The same result can be achieved by a dynamic arrangement, that is to say, by causing a current of silicon tetrachloride, if desired, in admixture with hydrogen, to flow first over highly purified and highly heated silicon in order to form silicon dichloride, and then reacting the gaseous mixture of silicon dichloride and silicon tetrachloride at a somewhat lower temperature with the graphite body.

The following examples illustrate the invention:—

EXAMPLE 1

A rod shaped piece of silicon having a length of about 5 cm and a diameter of about 1 cm is arranged coaxially within a horizontal quartz tube having a diameter of about 40 mm. The piece of silicon is out of contact with the wall of the tube. At a distance of 0.5 to 1 cm from the piece of silicon, is placed a desiliconised silicon carbide crystal which is to be converted into a highly pure silicon carbide crystal. The piece of silicon is heated by high frequency electrical energy to 1380—1400° C. and the desiliconised silicon carbide body to 1300—1350° C. When the temperatures are constant a mixture of 4—6% by volume of silicon tetrabromide or silicon tetrachloride in argon is passed under reduced pressure of about 0.75 atmosphere at a velocity of 1—10 cm per second first over the heated silicon and then over the body to be siliconised. The silicon tetrahalide may be wholly or partially replaced by pure halogen or hydrogen halide. The silicon is carried along with the gas stream and the desiliconised body is reconverted into a coarsely crystalline highly pure silicon carbide.

EXAMPLE 2

The arrangement used is the same as that described in Example 1, except that the silicon is placed in a boat of quartz glass and is at a temperature of 1450—1500° C. The body to be siliconised has a temperature of 1350—1400° C. The gas used is a mixture of 2% by volume of bromine and about 3% by volume of hydrogen bromide in argon. In this case also the desiliconised body is converted into highly pure well formed silicon carbide crystals.

WHAT WE CLAIM IS:—

1. A process for the manufacture of shaped bodies of silicon carbide, wherein a shaped body of graphite, which may contain silicon carbide, is heated in a gaseous atmosphere comprising a silicon subhalide to convert the graphite into silicon carbide.

2. A process as claimed in Claim 1, wherein the heated shaped body of graphite is treated in a gaseous atmosphere comprising a silicon subhalide and hydrogen.

3. A process as claimed in Claim 1 or 2, which is carried out at a temperature in the range of 1300°—1400° C.

4. A process as claimed in any one of Claims 1—3, wherein there is used as the graphite shaped body one which has been obtained by wholly or partially removing the silicon from a shaped body of commercial silicon carbide by treating the latter with chlorine or a silicon tetrahalide at a temperature above 1000° C.

5. A process for the manufacture of shaped bodies of silicon carbide, conducted substantially as described in either of the examples herein.

6. Shaped bodies of silicon carbide whenever made by the process claimed in any one of Claims 1—3.

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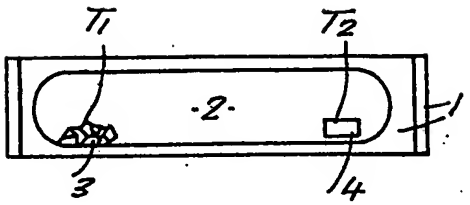
1 SHEET This drawing is a reproduction of the Original on a reduced scale.

Fig. 1.



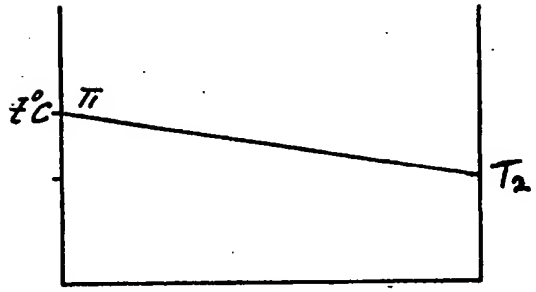


Fig. 2.